Research Needs and Opportunities for Characterization of Activated Samples at X-Ray and Neutron User Facilities

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[Appropriate cover art]

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1. **Executive Summary**

A new generation of science-based models and high performance simulations, coupled with credible uncertainty quantification, could reduce the time and cost required for nuclear plant licensing and fuel qualification. Because the complementary and necessary irradiations and characterization of activated specimens are expensive, the intelligent application of innovative experiments is important to realizing the potential savings. X-ray and neutron facilities are relevant because they provide measurement capabilities over wider time and length scales than typically are available in lab scale testing and have been fruitfully applied to wide range of materials challenges. However realizing the potential of these large-scale user facilities will require cultural changes at many facilities because of the requirements and infrastructure associated with safe handling of activated material. The provision of instrumentation and procedures necessary for efficient handling include sample preparation areas. Bringing to bear the full range of diffraction and spectroscopic techniques on smaller and smaller samples will be crucial. Particular areas of opportunity include interfaces and surface properties. Ultimately exciting scientific rewards can be realized by making measurements *in situ* in a radiation environment.

2. Introduction

Since the current generation of nuclear power reactors were designed nearly fifty years ago, dramatic advances in tools for the pursuit of materials science have been realized, ncluding in the arenas of X-ray and neutron user facilities and super-computer modeling. Using these tools it seems likely that break through insights will be possible in the next decade concerning the propensity for damage in materials exposed to high energy particles that introduce atomic displacement damage.

The field of radiation damage research is heavily influenced by its relevance to the safety case of engineering applications in fission power generation. Much of the research and development is directed at this end-use. Present fission reactors present many *managed* but poorly understood problems, concerning the strength and practical life of materials. Material selection in fission power applications is far from a mature technology. As higher burn-up, fluence and temperature for fission and ultimately fusion applications are considered the need for improved understanding is compelling. The potential societal implications are considerable for example in the United States alone, billions of dollars rest on the potential for lifetime extensions to be granted to an existing fleet of nuclear power stations.

The origins of radiation damage exists at the atomic scale but couple to the macroscale when engineering properties are compromised by the accumulation and interaction of radiation-induced defects. Ultimately the engineering performance of fuels or structural materials in a reactor depends on these defects and in particular on their interaction with the microstructure. At the atomic level the problems are as fundamental as any found in materials science. However extrapolating from atomistic insights to macroscopic lengthscales requires a complex and difficult synthesis of science, metallurgy and engineering.

This workshop was motivated by the potential that new tools at light and neutron sources have to proffer unique advances in our understanding of radiation damage. These measurements the have potential to inform and validate atomistic codes in ways that have hitherto been impossible. With wider application their insights might accelerate our understanding and certification of materials used in nuclear applications. A subordinate motivation was a desire to explore opportunities that would complement the Los Alamos “MaRIE” signature facility concept. The workshop was informed by three Office of Science workshops: Basic Research Needs for

Advanced Nuclear Energy Systems, Basic Research Needs for Materials Under Extreme Environments and Next Generation Photon Sources.

The workshop was held on September 20^{th} thru 22^{nd} of September 2009 and focused on user facilities. The charge addressed problems that warrant examination, current activity, opportunities that can be realized in the next few years and insights that could be realized by new diagnostics and experimental methods. The make-up of the approximately fifty attendees matched the scope of the scientific and engineering problem. Attendees came from many of the US National Laboratories including Los Alamos, Lawrence Livermore, Argonne, Oak Ridge, Pacific Northwest and Idaho. US industry was represented by two attendees from the Electric Power Research Institute and a representative from the Nuclear Regulatory Commission. There were six from US universities and six international attendees.

3. Measurement needs

3.1 Introduction

The range of irradiated and activated samples that can benefit from examination at user facilities is considerable. Opportunities span the range from fundamental studies to practical studies on fuels or structural materials over a range of dose, rate and temperature conditions. There are immediate issues such as fuel pool liners and future issues implicit in the design of a conceptual fusion reactor first wall. Noting the current advocacy for greater impact of modeling to certification and discovery, the meeting focused on tools that would contribute to the perennial desire to link models over comprehensive length and temporal scales.

In hot cells characterization tools typically span the range of lab scale techniques such as electron microscopy, Auger Atom probe spectroscopy, positron annihilation spectroscopy , Raman, mechanical testing etc. Often the studied materials are surveillance coupons and the properties of interest are those most pertinent to engineering: hardness, tensile properties, toughness, residual stress, texture, yield strength, strain hardening, corrosion & oxidation rates, and materials compatibility. Handling irradiated samples in hot cells with remote handling is routine if not cheap. Whereas X-ray and neutron user facilities often provide opportunities to examine a wider range of phenomena non-destructively on smaller samples with greater precision, spatial and temporal resolution this capability is sparsely applied in no small part because of the reticence and lack of infrastructure at most user facilities to handle highly activated materials. Nevertheless there are counter examples such as the Stanford Synchrotron Radiation Lightsource which accepts samples up to 10GBq (as indeed will the MARS soleil facility in France) or the Chalk River facility where shielded containers have been used to make neutron diffraction measurements on samples up to 20 000R/hour. Listed below are three problem areas that could strongly benefit from increased measurements.

3.2 Nuclear fuels

UO_2 and mixed (Uranium and plutonium) oxide are the primary candidates for fuels for new reactors over the next 25 years. The factors limiting fuel performance are the defect distributions, voids, bubbles, cracks, precipitates, new chemical phases, alloy species redistribution, recrystallization and grain growth generated by radiation. Typically the information is needed as a function of radial position from the centerline to the fuel cladding and has been expensively obtained by classical microscopy techniques but new methods may allow them to be determined non-destructively.

One of the potential advantages of fast-breeder fuels is their potential to offer higher burn-up when compared to current nuclear fuels (10% versus 3%). One candidate design is TRISO which comprises spheres (mm diameter)of UC surrounded by moderating layers of carbon, contained within a stainless steel cladding. The result is a fuel which is inhomogeneous on the scale of the size of the spheres. Therefore their behavior will also be locally inhomogeneous. Since they are expected to operate at higher temperatures, perhaps using liquid metal coolants, experiment capabilities are urgently needed to follow the inhomogeneous formation of defects, voids, crack, bubbles and phase-changes that will form during irradiation. Fuel deterioration due to cracking of fuel within the cladding is one phenomenon which decreases the effective thermal conductivity and limits the rating of the fuel. Thus a major contribution to our understanding could be achieved if it were possible to follow fuel damage in-situ. This is true for regular oxide fuels but will be even more important where the swelling will be exacerbated in fast reactor fuels. Equally there is a need to monitor waste-form stability for which the evolution of new phases and damage accumulation affects issues such as the barriers to leachability.

3.3 Structural components

Knowledge of the degradation of the mechanical properties of the structural materials used within a reactor is vital to the safety case. Changes in yield and ultimate tensile strength, embrittlement and loss of fracture toughness are all common. The fracture toughness, yield-point and temperature determine what length a crack has to be before it grows rapidly and destroys the component. The effect of irradiation on fatigue (the response to cyclic loads provided by temperature cycling or vibrations caused by water flow), creep (the gradual growth of structures at high temperature and under stress), and their interaction is not well known.

Residual stresses associated, for example, with girth welds in nuclear pipe-work such as nozzles close to the reactor pressure vessel are important since their failure could result in a loss of coolant event. Since their magnitudes, generated by the welding process are often unknown, these residual stresses often play a more important role in limiting lifetimes than the in-service stresses. Some assumption or knowledge about these residual stresses is needed to model crack propagation through welded material whose toughness has been reduced by radiation in order to obtain a damage-tolerant estimate of remaining life. Thus radiation assisted stress corrosion cracking is a very important to both boiling water and pressurized water reactors. Electric Power Research Institute (EPRI) has major national and international programs to investigate the effect particularly concerning the respective effects associated with modern practices when compared to the manual methods of yester-year. (The welds of interest are often complicated since they join body-centered cubic pressure vessel steels to corrosion resistant stainless steel and are sometimes thick, involving 3-6in. piping in which the welds are highly constrained by the pipe thickness).

Another important aspect of reactor safety is the integrity of the fuel cladding against breakage and release of fuel and fission products. Thus the phenomena associated with cladding failure are also of considerable importance. For zircaloy cladding the issues are corrosion, hydride accumulation, growth driven by intergranular strains and crystallographic texture and exacerbation by stress fields induced by manufacture or welding. This is of more concern for fast (rather than thermal) reactor fuels where increased burn-up leads to greater swelling and hence pressure on the cladding.

3.4 New Materials

New materials of possible relevance for structural applications have recently been identified from research on so-called Generation 3 and 4 materials. These include oxide dispersion strengthened steels, ferrite/bainite (P91) alloys and alloys of the form Ti_3AlC_2. In many cases they appear to have good strength and radiation resistance but have little or no operational record in representative extreme radiation environments. Thus a scientific understanding of the interactions between strengthening features and the radiation induced defects is of paramount importance if they are to be certified for new applications. Segregation of alloying elements and the role of interfaces are two important areas of interest. Fundamental work is focused on the next decade but has immediate benefits if radiation resistant materials can be substituted in renovated reactors. Another identified need is to improve current materials within the envelope of their certification to avoid the drawn-out process of certifying anew.

It is becoming clear that some materials which have superior resistance to radiation, for instance the layered material CuNb, may attain this by virtue of the interfaces. Materials with strengthening features at the scale of nm often have superior mechanical properties. The reason for this improvement in properties may well be what happens in the interfaces rather than within the grains Cladding material shows void denuding near the grain boundaries suggesting that the latter, the interfaces, are sinks for defects. If interfaces and grain boundaries are playing a key role, then diffraction which originates in coherent effects in the grains may not be very helpful. Local probes sensitive to the conditions in grain boundaries are also called for. Fortunately, the size of synchrotron beams is becoming smaller while still retaining intensity. Within the next decade one may be able to focus on the diffuse scattering from interfaces such as grain boundaries.

4. Near term (1-5 year) opportunities

4.1 Methods for transformational insights

Much of what we think we know about primary damage formation comes from molecular dynamics simulations. However much experimental effort misses the spatial and temporal regimes most pertinent to MD by focusing on long range or bulk average phenomena such as resistivity, temperature, swelling , constitutive response and corrosion rates. Thus new tools which provide 3-D spatial distributions of defect and chemical distributions with atomistic resolution could prove invaluable. This is especially true if they have temporal resolution consistent with the phenomena of interest.

There is also a compelling need for engineering studies using new probes that can operate under extreme irradiation environments. For fuels, issues of interest include melt temperature as a function of actinide composition and chemistry; dimensional stability, thermal properties, and material diffusion as a function of chemistry, temperature and microstructure; heat generation from nuclear processes; fission product accumulation and gas release. Issues for cladding include strength and ductility as a function of microstructure, temperature, and chemistry; actinide and fission product diffusion; chemical reactions at fuel-clad interface. In either case the potential to make measurements during simulated failures such as loss-of-coolant accidents would allow failure margins to be explored.

Developments at 3rd and 4th generation light sources as well as at new neutron sources such as SNS or JPARC hold potential for unprecedented insights into fundamental processes that dictate radiation damage. Several specific areas of immediate opportunity were identified: diffraction measurements during loading of archival samples; residual stress measurements of structural welds; property determination of irradiated oxide dispersion strengthened / nano structured ferritic steels; inelastic neutron scattering on samples of atypical isotopic composition (e.g. Pu 242); resonant inelastic X-ray scattering fluorescence measurements (e.g. on Americium at high pressure) or elemental mapping on the sub-micron scale (0.03μm resolution) with a sensitivity to chemical species of parts per billion; and development of advanced X-ray and neutron focusing optics (e.g. for studies of ion beam irradiated layers).

Opportunities implicit in the Linac Coherent Light Source, LCLS, were a frequent focus. Its potential to observe with pico second resolution, defects, their interactions and dynamics may lead to better charting of damage pathways. The high-intensity short photon pulses offer experimental potential that, for the first time, complements high-performance computing atomistic models atteh shortest relevant spatial and temporal scales. Realizing this potential will require the development of techniques that can use such radiation sources to measure nucleation of defect clusters, bubbles/voids, image dislocations, grain boundaries and precipitates, and demonstrate the interaction of dislocations with defect clusters, bubbles and voids.

The subsequent sections address near term opportunities that would avail themselves of new radiographic and existing scattering tools available at existing user facilities

4.2 Radiography and tomography

The principal virtue of tomographic techniques is that they are non-destructive and can provide intermittent viewing of samples after exposure to damage or used in situ if the damage process is fast enough to warrant it.

X-ray tomography can be pursued using laboratory or synchrotron sources which complement one another by investigating components over a range of length scales. Laboratory tomography can effectively covers the spatial range of 1-10μm taking between ten hours and half an hour for a scan. It has been applied to components of the order of a millimeter in maximum dimension such as crack growth in nuclear graphite, damage in $LiTiO_3$ "pebbles" for fusion blankets and TRISO fuel elements. Defects in the SiC coating around a TRISO particle can readily be resolved. Conversely synchrotron tomography, carried out at major user facilities, and therefore requiring more organization, covers the size range 0.2-1.0 μm with scan times ranging between a few seconds and an hour. (High energy microtomography, with a resolution of 0.2 μm in a few seconds has been applied to viewing crack growth in compact tension samples in real time). Applications to visualizing defects in an irradiated microstructure are easy to envision.

Neutron tomography with a resolution of 200 μm continues to be limited by the availability of high resolution two dimensional detectors and by flux. Nevertheless new facilities are being built in Australia for examination of radioactive components and new general user facilities are being built at the ISIS neutron source in the UK and at the Spallation Neutron Source at Oak Ridge National Laboratory. At the SNQ continuous spallation source at the Paul Scherrer Institute in Switzerland it is used to examine large and highly radioactive samples. The advantages of a user facility for examination of active components at a licensed nuclear site are the simplification of transportation issues and the experience with handling the materials.

Diffraction contrast tomography allows a map to be made of the grains and their boundaries by correlation of the image of the grain in the x-ray transmitted beam and the diffracted Laue spot image of the grain. This tool has been used to map grain growth, grain boundary modification, recrystallization and phase changes. With micro-tomography on the same sample one can superpose the image of a propagating crack on the grain map to find out which grain boundaries are selected for crack growth. This is an immensely powerful tool with applications to defect distributions, voids and bubbles on the scale of the microstructure.

Phase contrast tomography is based on changes in refractive index and therefore phase at boundaries between regions of a sample with different densities. The technique often provides images where transmission based tomography is insensitive. The method has achieved successes in biology but has not yet been applied to activated materials.

4.3 Scattering

Small-angle scattering using X-rays and neutrons is common approach for studies of inhomogeneities covering size ranges from Å up to 10μm (using ultra small-angle techniques).

Small-angle neutron scattering measurements (SANS) and ultra small-angle scattering have measured the distribution of voids and defects in oxide dispersion strengthened steels by using the magnetic cross-section of the material to separate the non-magnetic defect scattering from other inhomogeneities in the steel in an unambiguous way. SANS has been also used to map creep cavitation in reactor pressure vessel steels close to the toe of a weld. There is now overlap (and agreement) between reciprocal space scattering methods of sizing particles and real space tomography in the 1-10 μm range.

Neutron diffraction measurements of residual stress have been made since the 1980's but the application to irradiated material is relatively new. Measurements of stresses in irradiated welds with 20,000R/hr on contact have been carried out at the Chalk River Nuclear Laboratories of Atomic Energy of Canada by the National Research Council of Canada staff. The welds were contained in specially designed containers such that the activity outside the container was 2-4mr/hr and therefore safe to deploy on the reactor main floor. The results, interestingly, showed a systematic reduction of residual stress with damage. Diffraction measurements of irradiated low-enriched uranium UMo fuel (75R/hr on contact) have also been made in shielded containers. The results showed that new phases appeared after irradiation plus an amorphous scattering component. A straightforward extension of the method would be to scan the fuel elements to find the distribution of phases as a function of position from the centre to the cladding generated by the temperature distribution from center to core.

Microdiffraction permits stress measurements to be made as a function of position within grains by the use of small x-ray beams generated by Baez-Kirkpatrick mirrors, with gauge volumes of order $0.4\times0.6\times0.7\ \mu m^3$. This allows the stresses to be mapped out across grains, around defects and dislocations and towards the grain boundaries. This is another powerful tool with good prospects of early application to understanding the stresses around bubbles and the genesis of cracks and crack fronts.

4.4 APS user survey

In November 2008 a survey gathered input on the needs of scientists planning to use synchrotron x-rays for examining activated materials in connection with the MR-CAT project at the Advanced Photon Source. Figure 4.1 (Slide 4 of Mei-mei Li's seed talk) shows the technique requirements which were identified in the survey. Small angle, ultra-small angle and diffuse scattering were considered important for examining defects. Both diffraction with mm size beams to measure phases and microdiffraction techniques to obtain stress distributions on the grain scale were identified. Surface diffraction and grazing incidence small angle scattering to examine near surface effects and corrosion were considered necessary. Tomography, radiography, phase contrast and fluorescence imaging were all identified as highly important. XAFS, micro-EXAFS and x-ray photoemission were considered to be highly valuable spectroscopies.

The beam size requirements were divided nearly equally between sizes less than $0.1\ \mu m^2$, between 0.1 and $1.0 \mu m^2$ and large beams for bulk measurements. Control over a wide range of temperatures was needed. Temperatures between 500-1000°C corresponding to high temperature reactors and fusion first walls, between room temperature and 500°C corresponding to thermal reactors, and cryogenic temperatures to minimize thermal diffuse (phonon) scattering when examining diffuse scattering, were needed. Mechanical facilities included the ability to carry out in-situ crack initiation and growth (stress and imaging) measurements, tension, compression and fatigue and internal pressurization. Structural alloys (30%) comprise the largest group of likely topics followed by nuclear fuels (20%), transmutation products and actinides (20%) and SiC, oxides, nitrides and carbides (TRISO fuels) (30%). Requirements for specimen preparation, mounting, polishing and cutting were also identified.

5. New tools of decadal scope

5.1 Requirements in a decadal future facility

The new x-ray facilities currently under development, for example the free-electron laser, exceed the brightness of current sources by a wide margin so the likelihood of being able to probe even smaller volumes over smaller time intervals than at present is inevitable. On the second day of the workshop the participants considered what capabilities might be required in a facility that could juxtapose irradiation capability with advanced probes a decade in the future. Although there was little consensus on the of what it would take for science based certification to supercede the existing “cook and look” paradigm there was consensus that greater application of the neutron and X-ray sources for characterization purposes would be valuable. The capability to irradiate under a variety of conditions (fission reactors, ion beams, spallation sources, etc.) was considered essential. Material test reactors, spallation sources and ion beams were all discussed.

Regardless of the irradiation source, desirable functionality in a decadal future facility included in situ and ex situ measurements of a range of phenomena, e.g., diffraction during loading; examination of individual grains in activated samples; handling and characterization of “large”components; in situ creep properties with helium; defect kinetics measurements with resolution than overlap with models and spent fuel characterization characterization of spent fuels. Engineering requirements will likely be dominated by need for insight, particularly under high burn up conditions, on new fuel types such as Triso, MOX, or Thoria, of which we have comparatively little experience.

5.2 Common scientific needs

Breakthroughs in understanding in the next ten years are likely to come from the coupling between theory and modeling with experiment. At the experimental level this might call for designing experiments over the whole range of length scales and time scales with input from the modelers. At the facilities level, this calls for complementary techniques, such as irradiation facilities, modern examination methods using high intensity x-rays and neutrons, the means to handle the active materials, mechanical testing and chemistry related activities and the input of theorists and modelers. The MR-CAT project at APS illustrates the value of complementary techniques brought to bear on a single sample at the same time and ably supported by theory.

The major point is that the new non-destructive methods of examination can establish the bulk behavior and the development of the accumulating damage over time. Previous methods provided a highly local view but not the statistical behavior nor the time development. From the point of view of developing models of the defect structure the time development is vital. We have a theoretical picture of the formation of the initial cascade and the clustering that ensues over a short span of time based on kinetic Monte Carlo methods. However, how the clustering leads to a distribution of voids in space and in time and how these diffuse through the structure to develop a steady-state spatial distribution is not known. This covers a much longer time scale and needs to be followed. Since similar processes operate throughout the bulk of the material these may be followed with the aid of repeated x-ray and neutron diffuse scattering methods which sample the bulk in a statistical fashion.

For UO_2 fuels a thermal neutron environment is required since this is the situation in present day reactors. One can envisage making repeated ex-situ measurements on the same fuel pin after intervals of irradiation to observe the development of the voids and bubbles with time. One can also envisage making repeated x-ray and neutron diffraction measurements of the phase content of the pin as a function of distance from the center of the pin and as a function of time. One may be able to make diffraction contrast tomography measurements to find the grain morphology and examine recrystallization as a function of radial position and time if the gamma background problem can be circumvented. The question of in-situ examination of fuel by x-rays within the radiation source has been mooted. Undoubtedly penetrations can be made into the source to allow x-ray beams to enter and leave. However, high energy x-ray diffraction to follow the development of new phases under irradiation is within the realm of possibility. Likewise it may be possible to examine TRISO fuels, although this requires a neutron environment tailored to resemble a fast reactor. Repeated measurements taken in-situ and ex-situ of irradiated fuel would be feasible using tomography, diffraction to identify new phases and their distribution and grain configuration changes. These measurements would show the development over time of the voids and bubbles within the fuel and their distribution. Diffuse scattering would be difficult because of the inhomogeneous content of the element as well as its cylindrical shape.

It is desirable to do fatigue measurements within the radiation environment. Apart from the effect of embrittlement on crack growth, if the time scale for relaxation matches the time scale for fatigue, there will be a marked effect of one upon the other. Likewise it is important for fast reactors, where the operating temperature is higher than thermal reactors to determine whether high temperature creep affects the fatigue properties of structural components. There is a remote possibility that the high temperature creep may anneal out the effect of fatigue. Fatigue measurements require penetrations into the radiation source to transmit the cyclic force to the sample, radiation insensitive strain gauges and a fair amount of space but are possible. It would be relatively easy to take the fatigued and irradiated samples from the neutron source and examine the samples by the new tools to look for void clustering, crack growth and make measurements of stress around the crack tip to enable the theory of crack growth. For new materials the same considerations apply: the need to apply tensile and cyclic loads under irradiation and at a variety of temperatures to follow the creep-fatigue interaction. This would then be followed by examination by the new non-destructive tools.

5.3 User facility considerations

Considering the needs described above it is possible to consider what might be needed in a new facility. With the tools at hand, the easiest measurements are those on length scales close to that of the microstructure (1-100μm) where the time developments are slower. They are of direct engineering relevance. The faster length scales having to do with the physics of cascade formation are far harder to access with neutron irradiation than ion bombardment.

Thus the basic experimental building blocks are a neutron irradiation facility, and the possibility of in-situ examination by high energy x-rays. No less important are handling facilities enabling tensile testing and fatigue testing of engineering samples in hot cells, the ability to employ all the methods using x-rays from an intense source on highly active samples and easy access to neutron scattering tools also adapted for highly active samples. One also needs to have all the classical post irradiation microscopy such as electron microscopy. For example interpreting diffuse scattering data can be problematic if there are several sources of scattering since the particular kind of defect cannot be identified without complementary examination.

An extensive program of irradiation at LANSCE was carried out during the Accelerator Production of Tritium (APT) program. Irradiation with a spallation source adds a high-energy tail to the neutron spectrum compared with a fission reactor. In addition the production of He^4 from α-particles is a couple of hundred times higher than at a reactor. In this respect the spallation spectrum begins to approximate to a fusion spectrum. Many structural materials, steels and Al alloys were irradiated and archived. A pulsed fast neutron source, the Materials Test Station, MTS, could be incorporated in the LANSCE accelerator complex. This would have an average intensity that is twice as high as the ATR with intensity in the pulse 10000 times higher. On the other hand the neutron spectrum has a high energy component compared with a thermal reactor spectrum and the He^4 content is about 100 times higher. Both the high energy tail and the He^4 modify the response of materials.

One challenge for a new facility is rapid impact and one of the problems with neutron irradiation facilities is their relatively slow damage rate. There is a need for accelerated testing. A very important part of the solution for accelerated testing is ion beam bombardment which can simulate neutron damage. Ion beam irradiation sources provides damage rates that are typically several orders of magnitude faster than any conceiveable neutron irradiation facility. Although they lack the penetration, fission fragments and helium production in prototypic neutron irradiations ion irradiation can serve as a powerful complement. By choice of the bombarding ion one can simulate self-ion damage (say Fe and Cr in stainless steel) or fission gas accumulation. In a triple beam facility material is bombarded with heavy ions, as well as H^- ions and He^4. Synergistic effects of the three beams on damage have been recognized. In the MR-CAT project at the Advanced Photon Source (APS) the intent is to bombard foil samples in a triple beam and examine the damage in-situ with a synchrotron beam. Mechanical testing of foil samples is feasible although scaling up to engineering test samples and larger components is not assured.

Although neutron techniques cannot compete with x-rays from the view-point of miniature beams their advantages primarily lie in the variability of the scattering lengths as a way to distinguish elements close in the periodic table, light elements in the presence of heavy elements and the use of the magnetic cross section when this is feasible. The other major advantage of neutron diffraction and scattering is that it is not so difficult to shield against the gamma background from the irradiated materials as tests on fuels and welds has shown. Residual stresses in irradiated components will continue to be relevant to the regulatory process. Dedicated spectrometers for neutron experiments, making use of SANS, diffraction and stress measurements on irradiated samples will be an important part of a new facility.

Open questions

In fact the spectrum is more akin to a fast reactor or a fusion reactor spectrum. It is actually an unsettled question whether the rate of displacement actually changes the behavior so that it gives an accurate picture of the outcome.

The major problem is that there will always be a strong source of background arising from the gamma activity of the irradiated samples themselves. This will cover the same energy range as the x-rays used to do the experiments and may prove to be an insuperable obstacle.

Question of whether emerging nano/microscale testing can provide relevant certification data. This question is similar to that of whether science based models, perhaps selectively validated under non representative conditions, can make substantial inroads towards certification, when compared to long term irradiation under representative conditions.

6. Cross cutting themes

6.1 Modeling

The needs of the modelers serve to define the requirements for experiments at each length scale. At the atomic level, what are the length and time scales for the fundamental cascades generating the damage? What are the length and time scales for these to come to equilibrium in the material? How are these entities situated in the microstructure and how do they interact with intrinsic defects, such as the grain boundaries which arguably act as sinks. In turn the microstructural information is needed to calculate the macroscopic properties of the fuel, such as thermal conductivity, temperature and swelling

From the current perspective of modeling, the mathematical description of the microstructure (length scale 1.0 to 100μm) is recognized as the critical link between the behavior at the atomic level and the behavior of the material at the macroscopic level. In fact this is also true of the understanding of the mechanical properties of materials in general. At the atomic level, Kinetic Monte Carlo methods can describe the distributions of vacancies and interstitials immediately after (30fs) a fission product cascade has been initiated. In the following 20ps these vacancies and interstitials are thought to coalesce into small clusters over a spatial volume of diameter about 3nm (0.003 μm). The outstanding fundamental proviso is the description of the interatomic potential for U and its 5f electrons is as yet unsatisfactory.

How the voids and clusters populate the grains of fuel material is currently addressed by the "phase- field" model which replaces every grain boundary by an order parameter varying continuously from 0 to 1. The model can describe the genesis of gas bubbles and how they are distributed through the grains, in particular the experimentally observed denuded zones close to grain boundaries where there are fewer bubbles. With this description of the inhomogeneities within the grains, a constitutive law for the local thermal conductivity can be derived. In turn, finite element models of behavior on the macroscopic length scale use this information to calculate the average thermal conductivity as a function of position and hence derive temperature distributions through the fuel. This is one example of the direction of development of the models. While the modeling effort is close to connecting over the whole length scale, the field is not so much limited by computing power as by descriptive algorithms to describe behavior at the microstructure length scale. Another success has been the microstructural evolution of Fe0.9%Cu after irradiation. The initial vacancy-Cu clusters on a 10Å scale and their coalescence into a series of Cu precipitates have been predicted.

Currently many models are not hardware limited, but algorithm limited in developing physically-validated methodologies for predicting for example microstructural evolution or tensile stress-strain behavior using mesoscale (dislocation dynamics) models. For this reason much debate focused on measurements pertinent to atomistic and molecular dynamics calculations which were considered most in need of validation. A second theme pertained to the engagement of theorists. There was some belief that, even with new validation opportunities, the promise implicit in new generations of models would not be realized without complementary investment in what was called a "virtual computational end station". Essentially this theme advocated significant investment in developing the computational infrastructure necessary for the community to take advantage of insights that could be achieved by bringing light and neutron source characterizations to bear on activated materials.

6.2 Engagement with nuclear power industry

Interestingly, a stated opinion from the nuclear industry at the utility level is that there are no technical challenges for coping with irradiated material but that better allocation of funds is needed to support present examination methods. This suggests that there is currently a mis-match between what is seen as important within the nuclear industry with respect to post-irradiation examination and what researchers outside industry perceive. This mismatch should be addressed. Another suggestion was the alignment of goals of research workers outside the industry with the research topics identified by the Electric Power Research Institute in their interactions with the industry.

During breakout discussions, three broader themes arose that merit attention in any exploration of advanced measurement capabilities. First, some difficulty in engaging nuclear in the advocacy of long term research was noted. The general assumption was that short-term commercial imperatives focused industry attention on the immediacy of existing light water reactor recertifications. The participants spent some time discussing this issue with solutions ranging from more aggressive participation of the research community at EPRI and NRC meetings; education of the user communities on the way safety case certification occurs now; definition of a DOE champion; efforts to bridge the gaps between the different modus operandi of the Office of Science and the Office of Nuclear Energy; and addressing the issue of conflict of interest and intellectual property issues.

7. Conclusion

If the promise of the so-called nuclear renaissance is to be realized, especially in the United States, the breadth and depth of the nuclear science and engineering community must be enhanced substantially. In particular there is a need to revitalize the materials science of radiation damage. It was clear to workshop participants that x-ray and neutron sources at national user facilities have an important role to play in this endeavor. Further, in addition to cultural changes to allow the full exploitation of currently available tools and techniques, new capabilities need to be developed if science-based certification is to play a role in the resurgence of nuclear energy. Finally, given the magnitude and urgency of the need for carbon-neutral energy, approaches must be found to reduce the time and cost associated with licensing and certification.

4.4 Present fast-neutron sources

The Advanced Test Reactor (ATR) at Idaho National Laboratory is a user facility for neutron irradiation and subsequent examination. It offers a very high flux of fast neutrons (5×10^{14}n/cm^2/s), new post-irradiation and rabbit facilities and a TRIGA reactor for neutron radiography. The ATR functions as the center-piece of an academic and industrial group and is also associated with the MR-CAT facility at the APS.The high flux reactor (HFIR) at Oak Ridge offers two locations for fast-neutron irradiation. An extensive program of irradiation at LANSCE was carried out during the Accelerator Production of Tritium (APT) program. Irradiation with a spallation source adds a high-energy tail to the neutron spectrum compared with a fission reactor. In addition the production of He^4 from α-particles is a couple of hundred times higher than at a reactor. In this respect the spallation spectrum begins to approximate to a fusion spectrum. Many structural materials, steels and Al alloys were irradiated and archived.

Appendix A:- Workshop Agenda

Research Needs and Opportunities for Characterization of Activated Samples at X-Ray and Neutron User Facilities

Sunday 20th September 2009			
5:30 p.m.	Registration		Tesuque B, C
6:30 pm	Welcome	Tom Holden (NST)	Tesuque B, C
6:40 pm	Overview of DOE Workshops	Mike Fluss (LLNL)	Tesuque B, C
7:00 pm	MaRIE, a proposed signature facility	John Sarrao (LANL)	Tesuque B, C
7:30 pm	Free time		
Monday 21st September 2009			
7:30 am	Continental breakfast		Portal
8:00 am	Workshop goal	Tom Holden (NST)	Tesuque A,B
8:05 am	Workshop philosophy & Rationale	Mark Bourke (LANL)	Tesuque A,B
Session 1:- Measurement needs for activated samples – customer focus	Chair/Scribe	Rick Holt / Peter Hosemann	
8:30 am	Developing advanced nuclear structural materials for use in high radiation environments: An Australian perspective	Lyndon Edwards (ANSTO)	Tesuque A,B
8:55 am	Microstructure Modeling of Nuclear Fuel	Dieter Wolf (INL)	Tesuque A,B
9:20 am	Welding residual stress and material property measurements on irradiated	David Rudland (NRC)	Tesuque A,B

	materials		
9:45 am	Breakout # 1 objectives	Jack Shlachter (LANL)	Tesuque A,B
10:00 am	Refreshments		Portal
Breakout # 1 -- In parallel			
Subgroup 1 *Regulatory & Licensing* Chair / Scribe James Wall / Heather Volz			
10:15 am	Seed Talk	Tiangan Lian (EPRI)	Tesuque A
10:25 am	Address breakout #1 charge	Subgroup	Tesuque A
Subgroup 2 *Industry* Chair/Scribe Phil Withers / Don Brown			
10:15 am	Seed Talk	Rick Holt (Queens University)	Tesuque B
10:25 am	Address breakout #1 charge	Subgroup	Tesuque B
Subgroup 3 *Modeling* Chair/Scribe Brian Wirth / Turab Lookman			
10:15 am	Seed Talk	Malcolm Stocks (ORNL)	Tesuque C
10:25 am	Address breakout #1 charge	Subgroup	Tesuque C
12:00	Lunch		
12:30 pm	Research on Transuranium Systems: Present Capabilities with Large Central Facilities (Lunchtime talk)	Gerry Lander (ITU retd.)	
1:30 pm	Breakout #1 chair summaries		Tesuque B,C
Session 2 Ongoing measurements on activated samples Chair/Scribe Angus Lawson / Heather Hawkins			
2:00 pm	Neutron, Synchrotron & Laboratory X-ray imaging of Active Samples	Phillip Withers (UMIST)	Tesuque B,C
2:25 pm	Radiation Damage Effects in Structural Materials using Swiss Spallation Neutron Source and Light Source Facilities	Yong Dai (PSI)	Tesuque B,C
2:50 pm	Radiation Damage Studies at Los Alamos Neutron Science Center (LANSCE) ; What We Did and What we would Still Like to Do	Walt Sommer (LANL Retd.)	Tesuque B,C
3:15 pm	Breakout # 2 objectives	Jack Shlachter (LANL)	Tesuque B,C

3:20 pm	Refreshments		
Breakout # 2 --- In parallel			
Subgroup 1 *Length scales - Nanometers (defects)* Chair / Scribe Mike Nastasi / Thomas Proffen			
3:40 pm	Seed Talk	Jim Stubbins (UUC)	Tesuque A
3:50 pm	Address breakout #2 charge	Subgroup	Tesuque A
Subgroup 2 *Length scale - Microns (Microstructure)* Chair / scribe Gene Ice / Steve Conradson			
3:40 pm	Seed Talk	MeiMei Li (ANL)	Tesuque B
3:50 pm	Address breakout #2 charge	Subgroup	Tesuque B
Subgroup 3 *Length scale – mm (components)* Chair / Scribe Jeff Terry / Don Brown			
3:40 pm	Seed Talk	Ron Rogge (Chalk River Can.)	Tesuque C
3:50 pm	Address breakout #2 charge	Subgroup	Tesuque C
6:00 pm	Free Time		
7:00 pm	Dinner		
8:00 pm	Material & Fuel Testing for Advanced Nuclear Systems (After dinner talk)	Paul Lisowski (DOE Office of Nuclear Energy)	
Tuesday 22nd September 2009			
7:30 am	Continental breakfast		Portal
8:00 am	Breakout #2 chair summaries		Tesuque B,C
Session 3:- New tools and new facilities decadal scope Chair Mike Fluss Scribe Ming Tang			
8:30 am	Materials By Design	Marius Stan (LANL)	Tesuque B,C
8:55 am	Light source opportunities	Gene Ice (ORNL)	Tesuque B,C
9:20 am	Facility needs	Rick Kurtz (PNNL)	

9:45 am	In situ measurements in a radiation environment	Mark Bourke (LANL)	Tesuque B,C
9:55 am	Breakout # 3 objectives	Jack Shlachter (LANL)	Tesuque B,C
10:00 am	Refreshments		
Breakout #3 in parallel			
Subgroup 1 *structural components* Chair / Scribe Randy Nanstad / Tiangan Lian			
10:20 am	Seed Talk	James Wall (EPRI)	Tesuque A
10:30 am	Address breakout #3 charge	Subgroup	Tesuque A
Subgroup 2 *Fuels and Waste* Chair / Scribe Ken McClellan / Rex Hjelm			
10:20 am	Seed Talk	Akos Horvath (HAS KFKI)	Tesuque B
10:30 am	Address breakout #3 charge	Subgroup	Tesuque B
Subgroup 3 *Fundamental Materials* Chair/Scribe Alan Hurd / MeiMei Li			
10:20 am	Seed Talk	Brian Wirth (UCB)	Tesuque C
10:30 am	Address breakout #3 charge	Subgroup	Tesuque C
12:00 pm	Lunch		
12:30 pm	Advanced Test Reactor national scientific user facility (Lunchtime talk)	Mitch Meyer (INL)	
1:30 pm	Breakout #3 chair summaries		
Session 4:- Ten Year Outlook - Panel discussion Chair / Scribe Gerry Lander / Stuart Maloy			
2:00 pm	Panel discussion	Rick Holt (Queen's Univ.) Jim Stubbins (UUC) David Rudland (NRC) Rick Kurtz (PNNL) Dieter Wolf (INL)	
3:00 pm	Open discussion	All	
4:00 pm	Refreshments		

Session 5:- Synthesize Draft Conclusions Chair Mark Bourke			
4:20	Draft Workshop Conclusions	Lyndon Edwards (ANSTO)	
5:00 pm	Written input	Tom Holden (NST)	
5:30 pm	Close		
7:00 pm	Dinner		
8:00 pm	Nonproliferation considerations for nuclear energy and nuclear weapons (after dinner talk)	DV Rao (LANL)	

Appendix B :- Participants and Readers

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